

# Light Emission in Silicon from Carbon Nanotubes

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## Abstract

The use of optics in microelectronic circuits to overcome the limitation of metallic interconnects is more and more considered as a viable solution. Among future silicon compatible materials, carbon nanotubes are promising candidates thanks to their ability to emit, modulate and detect light in the wavelength range of silicon transparency. We report the first integration of carbon nanotubes with silicon waveguides, successfully coupling their emission and absorption properties. A complete study of this coupling between carbon nanotubes and silicon waveguides was carried out, which led to the demonstration of the temperature-independent emission from carbon nanotubes in silicon at a wavelength of  $1.3\ \mu\text{m}$ . This represents the first milestone in the development of photonics based on carbon nanotubes on silicon.

**Keywords:** nanotube, photonics, silicon, photoluminescence

Enhancing microprocessor performances is becoming increasingly more complex. This complexity stems from the rising transistor count and the transistor's shrinking size in the quest to follow Moore's Law.<sup>1</sup> As a consequence, power consumption in microprocessor increases, and the on-chip communication between different components becomes more and more difficult, and

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is even now a key-point for future multi-core generations.<sup>2</sup> To overcome these problems, one of the most promising solutions is the use of optical interconnects, which combine high data rate transmission, low power consumption, synchronization and crosstalk.<sup>3,4</sup> In recent years, silicon photonics was extensively studied for the realisation of high-speed optical links.<sup>5-7</sup> Nowadays, compact photonic structures are achieved due to the strong refractive index contrast between silicon and silica and low loss optical propagation in the wavelengths range from 1.25  $\mu\text{m}$  to 1.65  $\mu\text{m}$ . In particular, various passive devices for wavelength multiplexing or light distribution (90° bends...) are easily feasible with silicon photonics technology.<sup>8,9</sup>

Nevertheless, silicon is an indirect bandgap material, and its optoelectronic properties are insufficient to generate light and not sensitive enough to detect the flux of photons transmitted in an optical link. Other materials, such as III-V semiconductors, are good alternatives for light emission,<sup>10,11</sup> while germanium could realize high speed photodetectors.<sup>12,13</sup> Silicon is still used for efficient and high speed optical modulators.<sup>14</sup> The integration of all these materials on silicon is technically possible, but as different and sometimes non-compatible process are used, the resulting scheme is not cost-effective, and consequently reduce the use of silicon photonics for a broad application domain. A monolithic integration of laser source, optical modulator, and photodetector with a common material would be much more favorable for emergence of photonics.

We envision the use of carbon nanotubes for all active optoelectronic devices in silicon in order to avoid these non compatible processes. Carbon nanotubes (SWNT) are a very versatile material, presenting at the same time very good electronic properties,<sup>15</sup> and also optical properties.<sup>16-18</sup> They display strong photo- and electro-luminescence, in the 1-2  $\mu\text{m}$  wavelength range,<sup>19-22</sup> and the emission could be tuned by selecting a precise nanotube diameter and chirality.<sup>23</sup> The possibility to use electrical pumping for luminescence generation is extremely interesting for the realization of electrically pumped optical laser.<sup>24,25</sup> Recent works revealed that SWNT displays electroabsorption properties, which could be used to achieve optical modulation.<sup>26,27</sup> Finally, nanotubes present various absorption bands in the 1-2  $\mu\text{m}$  range, allowing realization of photodetectors.<sup>28,29</sup> Therefore, carbon nanotubes are very good candidates to solve integration issues in

silicon photonics, and make cost-effective and reliable photonics. Moreover, a side advantage of the use of nanotubes for photonics is that the current research on the use of nanotube for nanoelectronics<sup>30</sup> will facilitate the integration between photonics and electronics.

In this paper, we propose a way to integrate nanotubes with silicon photonics technology, which is a key-point for future realisation of carbon nanotube based photonic devices. This work looks into the difficulties of coupling the optical properties of a 1D nanomaterial (SWNT) to a bulk 3D material like silicon. In this context, we report the first integration of nanotubes' absorption and emission properties in silicon waveguides at telecommunication wavelengths around  $1.3\ \mu\text{m}$ .

## Results and discussions

### Integration scheme design

The integration scheme considered is based on the insertion of an interacting zone in an input/output silicon waveguide. It is constituted of a sub-micron silicon waveguide embedded in a carbon nanotubes composite, as schematically presented in Fig. 1. The input and output waveguides have both a width  $W_g$  of 450 nm. For such geometry, the optical mode is strongly confined into the waveguide (Fig. 1A) due to the high refractive index contrast between silicon (optical index 3.45) and its top and bottom cladding layers where the index is near 1.45. This confinement prevents strong interactions with the surrounding media, allowing high transmission of optical signal. On the other hand, the waveguide in the interaction region with carbon nanotubes has a width  $W_i$  below the critical minimum confinement width  $W_c$ , which is the limit under a significant fraction of the optical mode starts to leak outside the silicon waveguide. At  $1.3\ \mu\text{m}$ ,  $W_c$  is around 400 nm. A judicious shrinkage of the waveguide width  $W_i$  allows both deconfining a controlled fraction of the optical mode and preserving the propagation of the optical wave along the waveguide. Fig. 1B illustrates the mode confinement calculated for a  $W_i$  of 275 nm and shows the optical mode spreading outside the waveguide silicon core. As a significant fraction of the energy is propagating outside the waveguide, the guided mode will have a strong interaction with the active SWNT based

polymer top cladding layer. To minimize optical losses in the transition between the input/output waveguide ( $W_g = 450$  nm) and the interaction region waveguide ( $W_i = 275$  nm), adiabatic tapers are required (typically 300  $\mu$ m long).

## Technology

The substrate is a silicon-on-insulator (SOI) with a 220 nm thick silicon layer on top of a 1  $\mu$ m thick buried silica layer. Although details of the technological steps are given in supporting information, briefly, silicon waveguides were made using e-beam lithography patterning followed by reactive ion plasma etching (RIE). A 500 nm thick silicon dioxide ( $\text{SiO}_2$ ) protection layer was deposited onto the silicon wafer using a plasma-enhanced chemical vapor deposition (PECVD) technique. A silica recess of length  $L$  corresponding to the interaction region was etched down to the buried silicon oxide layer in order to expose the vicinity of the waveguide allowing interactions with the surrounding medium, and in particular SWNT. (1C).

The single-wall carbon nanotube (SWNT) film was prepared as follow: as-prepared HiPCO SWNT powder (Unydim Inc.) were mixed with poly-9,9-di-n-octyl-fluorenyl-2,7-diyl (PFO) in toluene at a ratio of SWNT (5 mg): PFO (5 mg): toluene (30 ml). This mixture was homogenized by sonication (for 1 h using a water-bath sonicator and 15 min using a tip sonicator) and was centrifuged for 5-60 min using a desktop centrifuge (angle rotor type, 10,000 g). Solution was then drop casted directly into the previously defined silica recess to form a 1  $\mu$ m thick layer. Samples were further annealed at 180°C for 15 min to improve optical quality of the SWNT/PFO film.<sup>19,31</sup> This layer was then delimited around the interaction region by another  $\text{O}_2$ -based RIE to remove the SWNT/PFO film on the undesired areas (1D).

The evolution of the optical mode fraction confined in the SWNT/PFO layer (filling factor  $\mathcal{F}$ ) was determined as a function of the waveguide width  $W$  by mode-solving numerical simulations using an home-made simulation code, and are displayed in 2A. The multimode/singlemode limit is found to be around 500 nm. As  $W$  narrows down, the filling factor (*i.e.* the amount of energy in the SWNT/PFO layer) increases, thus strengthening optical mode interaction with carbon nanotubes.

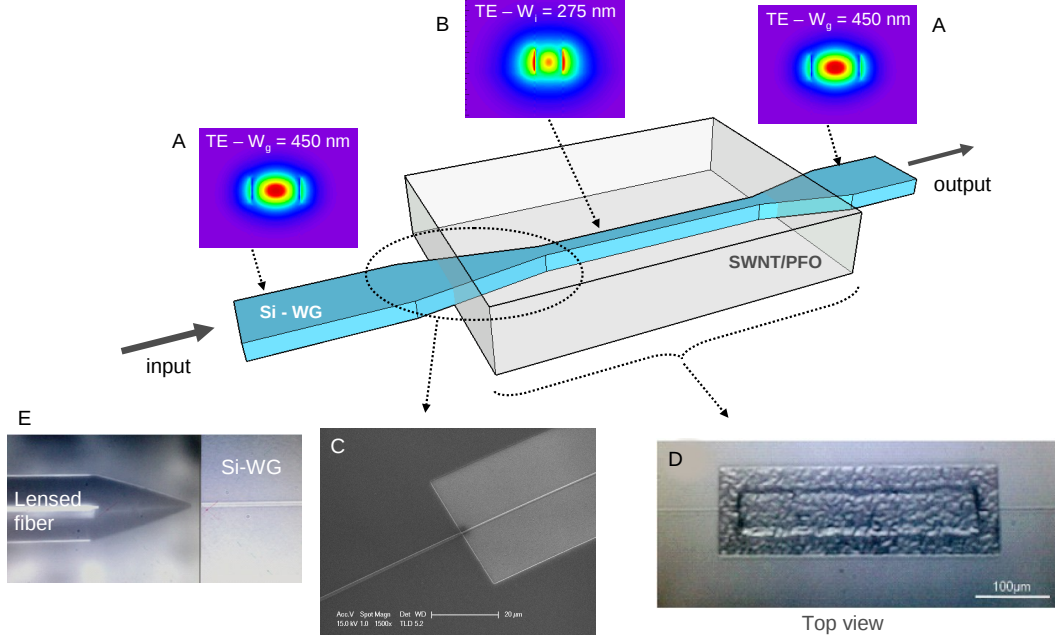


Figure 1: Integration scheme of carbon nanotubes thin layer composite with silicon waveguide. (a) Input and output single-mode silicon waveguides with a height of 220 nm and a width  $W_g$  of 450 nm. The optical mode for TE polarization is strongly confined into the silicon waveguide. (b) Interaction region with carbon nanotubes and the silicon waveguide, with a height of 220 nm and a width  $W_i$  of 275 nm. The guided mode is deconfined, and a significant fraction of the energy propagate outside the waveguide. (c) SEM view of the adiabatic taper which adapt the mode from the input waveguide ( $W_g$ ) to the interaction zone waveguide ( $W_i$ ). (d) Optical microscope top view of the final device, with the silica recess and SWNT/PFO thin layer on top of it. The apparent roughness on top of the SWNT/PFO thin film is due to the low temperature  $\text{SiO}_2$  hard mask used in the last technological step. (e) Optical microscope view of the lensed fiber used to inject the input light into the cleaved waveguide facet.

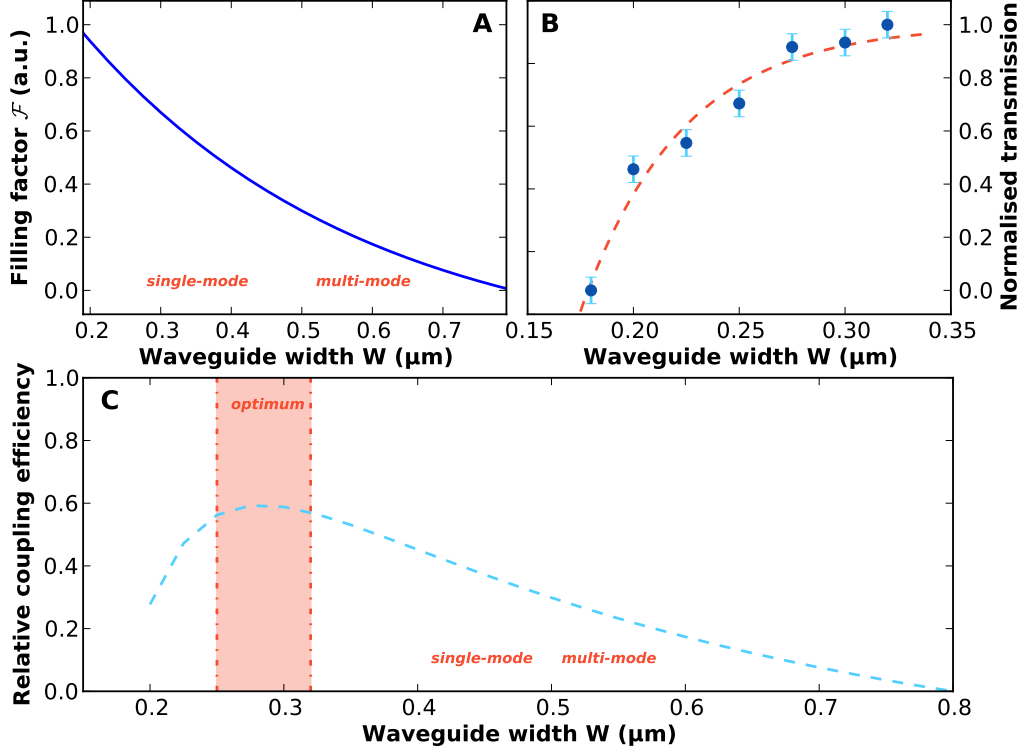


Figure 2: Determination of the waveguide width  $W$  to optimize the interaction between the optical mode in silicon waveguide and carbon nanotubes. (a) Simulation of the evolution of the optical mode fraction confined in SWNT/PFO layer (filling factor  $\mathcal{F}$ ) as a function of the silicon waveguide width  $W$ . (b) Experimental results of the normalized transmission as a function of the silicon waveguide width  $W$ . (c) Optimal conditions which fulfill low loss and strong interaction of the optical modes with SWNT. This results from the convolution of (a) and (b). The optimum waveguide width range is between 250 nm to 330 nm.

Ideally, the waveguide width  $W$  should be reduced down to 150 nm and even less to optimize light interaction with SWNT. However, silicon waveguides are on top of a 1  $\mu\text{m}$  thick  $\text{SiO}_2$  layer. If the optical mode transmitted into the waveguide is too deconfined, it will start to leak through the  $\text{SiO}_2$  layer towards the silicon substrate, resulting in huge losses. 2B displays experimental transmission results performed on optical waveguide of the same length with several width  $W$ . As  $W$  is reduced, the optical transmission through the waveguide exponentially decreases.

There is an optimum for a waveguide width to fulfill both high transmission and strong interactions with SWNT/PFO layer. This optimum could be determined thanks to the convolution of the experimental transmission and the simulated filling factor. The obtained result is displayed in 2C. The optimum waveguide width  $W$  was found to be between 250 and 330 nm. Consequently, a waveguide width  $W_i$  of 275 nm was used for the interaction region.

## Absorption coupling

The integration of SWNT in silicon and the coupling of their optical properties was first demonstrated by measuring the absorption of the upper SWNT/PFO layer across the waveguide. A laser beam from a tunable fibered laser source with an emission wavelength centered at 1.3  $\mu\text{m}$  was used at TE polarization (*i.e.* electric field parallel to the substrate). Input light was injected into the cleaved waveguide facet using a lensed fiber (1E). A liquid nitrogen cooled InGaAs monocal detector recorded the output beam through a monochromator.

3A reports the absorption spectrum of the carbon nanotubes through the silicon waveguide. In this case, the waveguide width  $W_i$  and length  $L$  in the interaction region were 275 nm and 400  $\mu\text{m}$  respectively. Several absorption peaks could be observed, each one corresponding to the specific absorption of one kind of nanotubes with specific  $(n,m)$  index. In the considered wavelength range (1.25-1.4  $\mu\text{m}$ ), three different SWNT are clearly identified, corresponding to (10,5), (8,7) and (9,7) indexes.

The influence of the interaction length  $L$  was studied from the absorption contrast  $C$  of the (9,7) SWNT defined as:

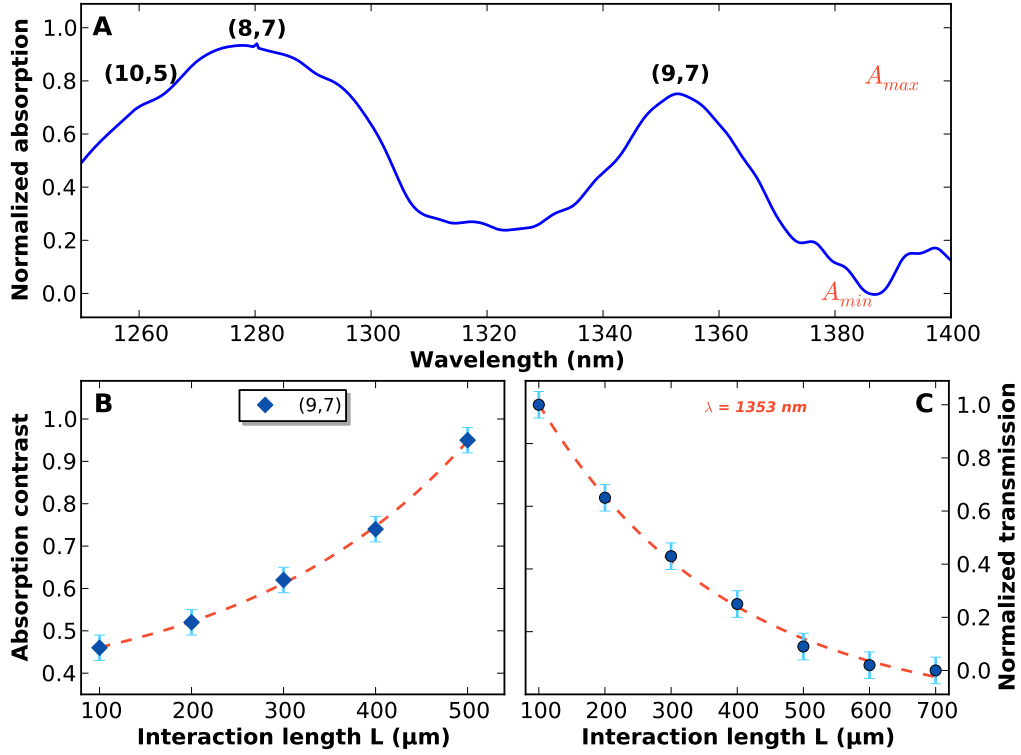


Figure 3: Carbon nanotube absorption measurements in silicon waveguide. Evolution with the interaction length  $L$ . (a) Normalized absorption as a function of wavelength from 1250 nm to 1400 nm. (10,5), (8,7) and (9,7) species are clearly observed. (b) Absorption contrast  $C$  of the (9,7) nanotube, defined by  $\frac{A_{max}-A_{min}}{A_{max}+A_{min}}$  as a function of the interaction length  $L$ . (c) Normalized transmission of the (9,7) nanotube at  $\lambda = 1353 \text{ nm}$  as a function of the interaction length  $L$ .



$$C = \frac{A_{max} - A_{min}}{A_{max} + A_{min}} \quad (1)$$

where,  $A_{max}$  and  $A_{min}$  are respectively the maximum and minimum absorption of the (9,7) absorption peak, as defined on 3A.

Both the absorption contrast and the output intensity level were determined for several waveguide interaction lengths  $L$ , increasing the ranging from 100 to 500  $\mu\text{m}$ . Results are displayed in 3B and 3C, respectively. For the same waveguide width  $W_i$  of 275 nm, the absorption contrast  $C$  increase when  $L$  increase. On the same time, we found out that the waveguide transmission at  $\lambda = 1353$  nm (corresponding to the absorption peak of the (9,7) nanotube) is decreasing when  $L$  increase. Both results follow a Beer-Lambert type exponential law, indicating that the nature of the interaction between the optical mode propagated through the waveguide and carbon nanotubes is, indeed, optical absorption and not diffusive in nature. This result is the first signature of an effective coupling between SWNT absorption and the optical mode transmitted by the silicon waveguide.

Considering integration of carbon nanotubes with future photonic devices, there is a trade-off between the interaction with nanotubes (absorption contrast) and the waveguides transmission level. Several figures of merit could be proposed depending on the relative weight between absorption contrast and the global transmission of the device, however, an optimum interaction length  $L_i$  would be in the range 400 to 500  $\mu\text{m}$ , leading to very compact and low loss SWNT based photonic devices.

## Light emission

In order to investigate whether or not the proposed integration scheme is promising for carbon nanotube based optical sources, the light emission from SWNT into silicon waveguides was studied. A laser diode emitting at a wavelength of 802 nm was used to optically pump SWNT, in particular the (9,7) nanotube population. The incident laser beam arrived from the top of the silicon die, and

was focused on the silica recess, where carbon nanotubes could interact with the silicon waveguide. The light outgoing from the output waveguide facet, placed few millimeters away from the interaction region, was collected using a 20X microscope objective with 0.35 numerical aperture. The spectrum is displayed in 4A.

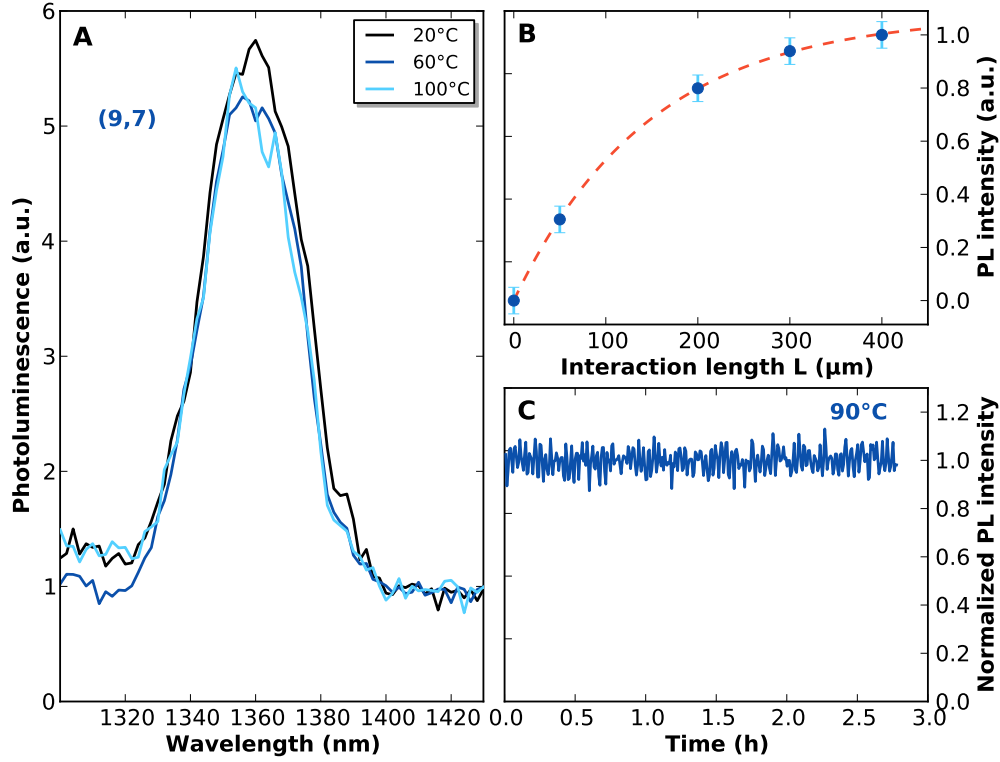


Figure 4: Coupling of carbon nanotubes PL in silicon waveguide. (a) Photoluminescence generated from the (9,7) nanotube under excitation by a 800 nm pump laser, coupled in silicon waveguide for three temperatures (20°C, 60°C and 100°C). No wavelength shift or signal reduction are observed. (b) Evolution of the photoluminescence intensity as a function of the interaction waveguide length  $L$ . (c) Time stability of the photoluminescence coupled in silicon waveguide at 90°C.

This (9,7) SWNT presents a strong photoluminescence (PL) peak, at a wavelength around  $1.35 \mu\text{m}$ , which is identical to the one reported in a thin PFO layer.<sup>19</sup> The influence of the interaction length  $L$  on the photoluminescence was also studied for several lengths between  $50 \mu\text{m}$  and  $400 \mu\text{m}$ . Results are displayed on 4B. For clarity, a point with an interaction length  $L = 0 \mu\text{m}$  and PL intensity of 0 was added to results. We noticed that the increase in PL intensity when the interaction length  $L$  increase is not linear. Indeed, its intensity increase until it reaches a maxi-

mum at  $L$  around  $400 \mu\text{m}$ . This evolution could be described by a Beer-Lambert type exponential law, meaning that the observed limitation in photoluminescence intensity is due to the SWNT/PFO layer secondary absorption. These results are consistent with previously obtained results for the absorption coupling and suggest that the optimal interaction length  $L_i$  for SWNT/PFO based photonic devices, relying on SWNT absorption or emission properties, would be around  $400 \mu\text{m}$ .

In order to furthermore determine the strength of carbon nanotubes for silicon photonic applications, we determined the SWNT emission stability with temperature. That is a major requirement for the achievement of most applications. Evolution of the (9,7) nanotubes emission through the silicon waveguide is presented in 4A for a given interaction length  $L_i$  of  $400 \mu\text{m}$ . We notice that SWNT PL peak did not shift in wavelength and keep the same intensity level for temperature ranging from  $10^\circ\text{C}$  to  $100^\circ\text{C}$ , which is typically the operating temperature of photonic and microelectronic systems. In order to assess thermal stability over time, PL intensity of (9,7) SWNT was recorded for 3 hours at  $90^\circ\text{C}$ , and is displayed in 4C. The emission through the waveguide remained constant, and no thermal degradation or intensity decrease was observed.

One important point in the coupling of the photoluminescence of a 1D material such as SWNT into the optical mode of a bulk 3D material like a silicon waveguide, is the coupling efficiency  $\Psi$ , which is the number of photon effectively transmitted through the waveguide over the total number of photon emitted by carbon nanotubes. That is:

$$\Psi = \frac{N_{\text{photons transmitted}}}{N_{\text{photons emitted}}} \quad (2)$$

However, this is a very difficult task to undertake in this case. Indeed,  $N_{\text{photons emitted}}$  is related to SWNT quantum yield  $\Phi$ , where:

$$\Phi = \frac{N_{\text{photons emitted}}}{N_{\text{photons absorbed}}} \quad (3)$$

Unfortunately,  $\Phi$  is not a well-known parameter in carbon nanotubes, and it may vary greatly, in the range  $10^{-4}$  to  $10^{-1}$ , depending of SWNT diameter, surrounding or length.<sup>32,33</sup> In particular,

$\Phi$  is not yet determined for PFO embedded SWNT.

In any case, it is possible to determine the *external effective coupling factor*  $Q_{eff}$ , where:

$$Q_{eff} = \Phi \cdot \Psi = \frac{N_{photons\ transmitted}}{N_{photons\ absorbed}} \quad (4)$$

Knowing the input and output power, the SWNT/PFO thin layer absorbance and the collecting setup transmission,  $Q_{eff}$  was estimated to be  $2 \cdot 10^{-5}$ .

If we estimate that carbon nanotubes quantum yield is low (*e.g.*  $\Phi = 10^{-4}$ ), this means that coupling efficiency  $\Psi$  is of the order of  $10^{-1}$ . In the future, the use of photonic crystals or slot waveguides might increase the coupling between carbon nanotubes and silicon waveguides.

In light of the results obtained so far, one could state that carbon nanotubes is a promising material for development of new compact and temperature independent photonics devices on silicon.

## Conclusions

In conclusion, the integration of carbon nanotubes properties (absorption and photoluminescence) in silicon waveguides was studied using an integration scheme based on evanescent waveguides. The coupling of carbon nanotubes photoluminescence into silicon waveguides has been demonstrated over a wide temperature range. The external effective coupling factor  $Q_{eff}$  was estimated about  $2 \cdot 10^{-5}$ . In the future, improved integration schemes based on photonic crystals or slot waveguides will be considered to further increase carbon nanotubes coupling with photonic waveguides in order to fully exploit their extraordinary optical properties to achieve efficient optoelectronic devices in silicon.

# Materials and Methods

## Carbon nanotubes

Single-wall nanotube powder was purchased from Unydim Inc. These nanotube were fabricated using the HiPCO process, and were not purified. poly-9,9-di-n-octyl-fluorenyl-2,7-diyl (PFO) was purchased from Sigma Aldrich. SWNT (5 mg) and PFO (5 mg) were mixed in toluene (30 ml). The mixture was homogeneized, first using a water-bath sonicator for 1h, second, using a tip sonicator at 20 % power for 15 min. In order to remove a part of metallic catalyst particles from the HiPCO process and other impurities, this mixture was then centrifugated using a desktop centrifuge (typically 5-60 min at 10.000 g). The supernatant is then collected and could be drop casted directly onto the photonics devices. The thin film are annealed at 180° for 15 min to improve their optical quality.

## Waveguide fabrication

Silicon-on-insulator (SOI) substrate were purchased from Soitec, with a top silicon layer of 220 nm, and a buried SiO<sub>2</sub> layer of 1  $\mu$ m. The technological steps for waveguide fabrication are as follow (see also Supplementary Materials S1):

- Step 1: SOI substrate preparation: Cleaning with acetone under sonication, followed by O<sub>2</sub> plasma cleaning.
- Step 2: Waveguides definition into the e-beam resist. Deposition of 300 nm thick MaN negative resist. E-beam patterning at 20 keV, 30 s resist developing in MIF 726.
- Step 3: Pattern transfert: Etching of silicon by RIE down to 6 nm thick silicon layer. RIE: SF<sub>6</sub> (20 sccm) and O<sub>2</sub> (5 sccm) at 30 W for 70 s.
- Step 4: Waveguide protection by SiO<sub>2</sub>: 500 nm thick SiO<sub>2</sub> layer is deposited on top of silicon waveguides at 300°C.

- Step 5: Interaction zone definition into the e-beam resist. Deposition of 300 nm thick positive ZEP resist. E-beam patterning at 20 keV. Developing: 30 s ZED50 / 30 s MIBK / 30 s IPA.
- Step 6: Interaction zone opening: Wet etching with diluted HF of silica down to silicon stopping layer. O<sub>2</sub> plasma for resist removal.
- Step 7: SWNT/PFO deposition: 1  $\mu\text{m}$  thick SWNT/PFO layer is deposited on the full die followed by a thermal annealing at 180°C.
- Step 8: SWNT/PFO protection with SiO<sub>2</sub>: 200 nm thick SiO<sub>2</sub> layer is deposited on top of the SWNT/PFO layer at low temperature (150°C) to prevent layer damage.
- Step 9: Definition of protected zone into the e-beam resist. MaN resist and e-beam patterning, developing.
- Step 10: SiO<sub>2</sub> etching using RIE : CHF<sub>3</sub> (50 sccm) and O<sub>2</sub> (3 sccm) at 325 W (12 mTorr) for 190 s.
- Step 11: Removal of SWNT/PFO outside defined zone by RIE etching. 50 % Ar and 50% O<sub>2</sub> at 300 W for 15 min.

## Simulations

Optical simulations were performed using an home-made 2D mode solver based on the full-vectorial finite-difference method<sup>34</sup> and focused on anisotropic dielectric waveguides.

The filling factor  $\mathcal{F}$ , measure of the fraction of the mode power flux in the SWNT/PFO layer, is defined by :

$$\mathcal{F} = \frac{\int^R P(s) ds}{\int^\infty P(s) ds}$$

where  $R$  is the SWNT/PFO layer and  $P(s)$  is the mode power flux.

## Determining $Q_{eff}$

Incoming power density on the silica recess  $J_i$  was estimated to be  $2.7 \cdot 10^6 \text{ W}\cdot\text{m}^2$ . A simple assumption is made considering the area around the waveguide whether a nanotube could interact or not. As the profile of the evanescent field is gaussian, it is considered that the whole energy of the optical mode is confined within  $5\sigma$  of the gaussian. That is, a nanotube inside  $5\sigma$  *may* interact with the waveguide, while a nanotube outside  $5\sigma$  *will not* interact with the waveguide. Consequently, only the SWNT/PFO thin layer surface inside this  $5\sigma$  limit will be considered. For a waveguide width  $W_i$  of 275 nm, the  $5\sigma$  limit is: 360 nm (cf. Supplementary Materials S2). That is, the surface area of the SWNT/PFO layer is  $S_{NT} = 1.1 \cdot 10^{-11} \text{ m}^2$ . The incident pump power on carbon nanotubes which could couple into the waveguide is  $P_i = J_i \cdot S_{NT} = 2.91 \cdot 10^{-5} \text{ W}$ . Absorption  $A$  of a 1  $\mu\text{m}$  thick SWNT/PFO layer at 805 nm is  $6.6 \cdot 10^{-2}$  (cf. Supplementary Materials S3). Absorbed power in carbon nanotubes  $P_a = A \cdot P_i = 1.92 \cdot 10^{-6} \text{ W}$ .

On the other hand, power of the collected photoluminescence from the waveguide was  $P_c = 1.27 \cdot 10^{-11} \text{ W}$ . The collecting setup losses were estimated to be 2 dB. So the transmitted power was  $P_t = P_c \cdot 10^{0.2} = 2.01 \cdot 10^{-11} \text{ W}$ .

The external effective coupling factor  $Q_{eff}$  could be determined by:

$$Q_{eff} = \frac{N_{photons \text{ transmitted}}}{N_{photons \text{ absorbed}}} = \frac{P_t/E_t}{P_a/E_a} = \frac{P_t}{P_a} \cdot \frac{\nu_a}{\nu_t} = \frac{P_t}{P_a} \cdot \frac{\lambda_t}{\lambda_a}$$

where  $E$  is the energy,  $\nu$  the frequency and  $\lambda$  the wavelength of the transmitted and absorbed photons.

Finally,  $Q_{eff} = 1.8 \cdot 10^{-5} \simeq 2 \cdot 10^{-5}$ .

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## Supporting Information Available

Detailed technological process of the waveguide fabrication (S1), evanescent mode profile of a 275 nm with waveguide (S2), and absorption spectra of a 1  $\mu\text{m}$  thick SWNT/PFO thin layer on silica (S3). This material is available free of charge *via* the internet at <http://pubs.acs.org>

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